

Coherent feedback from dissipation: the lasing mode volume of random lasers

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In any quantum or wave system dissipation leads to decoherence. Therefore, it was surprising in first instance when experiments on strongly lossy random lasers showed unambiguously by measurements of the photon statistics and of the lasing mode volume that coherent feedback is possible in such systems^{3,5,6}. In coherent-feedback lasers the photons form a far-from-equilibrium condensate in the sense that a single quantum state is occupied by a macroscopic number of photons¹⁴. We demonstrate that the lossy dynamics of random lasers alone imply a finite lasing mode volume, thus resolving the puzzle about coherent feedback without resonator. Our theory of random lasing including nonlinear gain and gain saturation predicts a characteristic dependence of this lasing mode volume on the pump intensity, which can be tested experimentally. The concept of a random laser (RL), i.e., lasing in a homogeneously disordered, laseractive medium without cavity, was introduced early on by Letokhov. It was then foreseen that light would be amplified by stimulated emission while diffusing through the random medium and covering an unlimited region in space. Consequently, a continuum of all the modes whose frequency lies within the laser transition line would participate in the lasing, leading to an uncorrelated (Bose-Einstein) photon number statistics (distributed feedback) and homogeneous laser emission from the entire system volume. By contrast, it was found in experiments on ZnO powder lasers that the laser light emission occurs from spatially strongly confined lasing spots with sharp, discrete emission lines and Poissonian photon number statistics, all three features being unambiguous signatures of coherent feedback, i.e. of laser emission from a single, spatially confined mode⁷. This spatial confinement is at the heart of understanding coherent feedback and the formation of a non-equilibrium photon condensate state in homogeneous random lasers. However, its origin remained obscure ever since its discovery, because the commonly known, possible confinement mechanisms cannot explain the observed experimental facts. Anderson localization (AL) of light in disordered media can be ruled out, because the photonic scattering mean free path ℓ is much longer than the wavelength of light λ , so that AL does not occur in the systems at hand, and the Ioffe Regel criterion is not met, i.e. $kl > 1$. Random microcavities, preformed accidentally in the disordered medium, are unlikely to be the origin

of coherent feedback, because neither would their intrinsic surface roughness allow for localized cavity modes to exist nor could they explain the observed dependence of the average lasing spot radius on the pump intensity^{3,5,6}.

Here we present a genuinely new mechanism for generation of a finite coherence or mode volume and coherent feedback, based on dissipation at the system surface. We show that optical losses at the surface imply that the light field in the bulk of the system can be coherent (correlated) only over a finite length ξ . Hence, this defines a volume within coherent feedback occurs, leading to Poissonian photon statistics. We identify ξ with the average radius of a lasing mode. It is dynamically generated by the highly non-linear, lossy dynamics of open laser systems and, as such, exists only in the lasing state. As we perceived, this confinement mechanism is closely related to the causality of both the propagation of light and the transport of light intensity. Moreover, this being a dynamical effect, we predict ξ to have a characteristic, namely power law, dependence on the laser pump intensity, which is found to be in qualitative agreement with available experiments^{3,5,6}.

As a RL system we consider specifically a layer of compressed powder of laser-active material, whereby the grains act as random scatterers and at the same time as amplifying medium. The layer has a thickness d and extends infinitely in the x-y plane, as depicted in Fig. 1a. The pump light covers a wide surface area and affects the entire layer, so that the pump intensity may be assumed homogeneous across the entire volume of the considered system. The laser material is characterized by an atomic four-level scheme with transition rates γ_{ij} as defined in Fig. 1b, although any other laser type would be possible.

In order to address the problem of the coherence volume in random lasers it is essential to set up the equation of motion for the coherent part of the radiation. This part is generated by stimulated emission only, although the quantum dynamics of the gain medium includes also spontaneous emission. The time evolution of the electric radiation field can be parameterized by as $\mathbf{E}(t, \mathbf{r}) = \mathbf{E}_0(t, \mathbf{r}) \exp[-i\omega t]$, where $\omega/2\pi$ is the light frequency and $\mathbf{E}_0(t, \mathbf{r}) \exp[-i\omega t]$ an amplitude function varying slowly on the scale of $2\pi/\omega$. We now observe that random lasers have to be treated as open systems, i.e., the loss-induced damping time is always much shorter than the lifetime γ_{12} of the upper atomic laser level (class B laser). In this strongly damped regime, the stimulated part of the polarization follows the electric field instantaneously. Hence, the wave equation for the stimulated emission is local in time and can be written in terms of the dielectric function $\varepsilon(\mathbf{r}, \mathbf{E}_0(t, \mathbf{r}))$ as

$$[\frac{\varepsilon(\mathbf{r}, \mathbf{E}_0(t, \mathbf{r}))}{c^2} \omega^2 + \nabla^2] \mathbf{E}(t, \mathbf{r}) = 0.$$

Note that $\varepsilon(\mathbf{r}, \mathbf{E}_0(t, \mathbf{r}))$ incorporates the full, nonlinear laser dynamics through its dependence on the field amplitude $\mathbf{E}_0(t, \mathbf{r})$ and depends on position \mathbf{r} both explicitly because of the random position of the dielectric scatterers

and implicitly through its dependence on \mathbf{E}_0 . The lasing state is characterized by a negative imaginary part of $\varepsilon(\mathbf{r}, \mathbf{E}_0(t, \mathbf{r}))$. It is a priori not known and therefore determined by solving a four level laser rate equation system (Siegmann) in steady state (see supplementary informations).

$$\begin{aligned}\frac{\partial N_3}{\partial t} &= \frac{N_0}{\tau_P} - \frac{N_3}{\tau_{32}} \\ \frac{\partial N_2}{\partial t} &= \frac{N_3}{\tau_{32}} - \left(\frac{1}{\tau_{21}} + \frac{1}{\tau_{nr}} \right) N_2 - \frac{(N_2 - N_1)}{\tau_{21}} n_{ph} \\ \frac{\partial N_1}{\partial t} &= \left(\frac{1}{\tau_{21}} + \frac{1}{\tau_{nr}} \right) N_2 + \frac{(N_2 - N_1)}{\tau_{21}} n_{ph} - \frac{N_1}{\tau_{10}} \\ \frac{\partial N_0}{\partial t} &= \frac{N_1}{\tau_{10}} - \frac{N_0}{\tau_P} \\ N_{tot} &= N_0 + N_1 + N_2 + N_3.\end{aligned}$$

The electronic transition rates from the ground state to the uppermost state and from the lower lasing level to the ground state are assumed to be large compared to all other transitions. Thus we focus on the description of the photon number n_{ph} and the inversion $n_2 = N_2/N_{tot} = \frac{\gamma_P}{\gamma_P + \gamma_{nr} + \gamma_{21}(n_{ph} + 1)}$ according to the Einstein rate equations in steady state. γ_P equals the pump rate, γ_{nr} represents the nonradiative decays and $\gamma_{21} = 1/\tau_{21}$ features the stimulated emission rate, which is the inverse of the lifetime or relaxation time of the upper lasing level. The resulting inversion of the occupation number thus defines the microscopic optical gain and the challenge is now to relate this gain to the coherent (correlated) part of the diffusing radiation. Therefore the laser rate system is coupled to sophisticated model for microscopic transport of light in disordered amplifying random media^{16?–18} based on Vollhardt-Wölfle theory for the transport of electrons¹², which allows for the consideration of the influences of the microstructure of the amplifying media. For the completeness of the description of random lasing it has been demanded earlier by Florescu et. al.¹⁴ that the effects due to multiple scattering of light and the internal Mie-resonances during these scattering events must be considered. We included those contributions by generalizing the diagrammatic description for the transport of light and we particularly focussed on the influence of interferences. Therefore we had to deal with a Dyson description of the propagation of the electromagnetic wave on the one hand and the Bethe-Salpeter-Equation for the propagation and dissipation of the energy density of light on the other. The complete description can be found in the supplementary, but to introduce the reader briefly to this fascinating subject, the Bethe-Salpeter-Equation for the two particle Green's function or in other words the energy propagator $\hat{\Phi}$ is given here - which looks a bit daunting at first glance but actually it is a treasure chest for the random laser theory.

$$\hat{\Phi} = \left(\hat{\mathbf{G}}^R \otimes \hat{\mathbf{G}}^A \right) \left[\mathbb{1} \otimes \mathbb{1} + \hat{\gamma} \hat{\Phi} \right].$$

The Bethe-Salpeter-Equation is the appropriate equation of motion for the coherent part of light intensity Φ . The Green's function \mathbf{G} itself describes the behavior of the electromagnetic wave, whereas the so called irreducible vertex $\hat{\gamma}$ reveals all interactions and the selfconsistence-loop is closed by the selfdependency of $\hat{\Phi}$. It has to be stressed that the propagation of light intensity in diffusive or strong scattering regimes obeys the second law of thermodynamics and thus the time scales of wave propagation and intensity propagation in fact can be separated. The results drawn from these considerations turned out to be really promising, because the diffusive processes, the interferences and localization processes can be clearly identified as separate contributions in the thereby derived diffusion constant $D(\Omega)$

$$D(\Omega) [1 - i\Omega \text{Re}\epsilon\omega\tau_a^2] = D_0^{tot} - \tau_a^2 D(\Omega) M(\omega).$$

The first term on the right hand side $D_0^{tot} = D_0 + D_b + D_s$ could be slightly overlooked on the search for interference effects which are represented through $M(\omega)$, the so called memory kernel. A detailed study reveals, however, that the memory kernel cannot cause true localization, i.e. $D = 0$, in laser active media, and D_0^{tot} cannot be compensated. D_0 reflects the bare diffusion, the dissipative renormalizations due to absorption and/gain in the system are summarized in the term D_b and D_s . After all we remark that this diffusion constant for systems, which are in any way laser-active, will never vanish, and therefore AL of light in such systems seems to be beyond the question in the discussion of random lasing.

From the microscopically derived correlation length ξ (see supplementary information) in steady state for strong pumping (see Fig. 4) one can draw several significant conclusions for the characteristics of a random laser

$$\xi = \frac{\alpha}{\sqrt{P}} + \xi_\infty$$

where α is a numerical constant and P the pumping. It is found that the natural antagonists, the pump strength P and the loss at the surfaces play a severe though in first instance counterintuitive role. The coherence length ξ in steady state clearly shrinks with the increase of the pump strength P whereas dissipation or loss at the surfaces of the depicted slab geometry (see Fig. 1) in contrary increases the correlation length. The meaning of both results become obvious when we go back to the roots of the idea of random lasing again. Dicke in 1968 proposed the possibility of the photon bomb by means of an infinite increase of the \mathbf{k} -modes in an amplifying media. Our results in contrast prove that the nature of the onset of lasing in amplifying random media is clearly described by

a finite correlation length and thus a finite mode volume which marks the exponential decay of the spatially and spectrally coherent light intensity inside the ZnO powder slab (Fig 3b). Loss at the surface however leads to an increase of the correlation length ξ . Both aspects, the dependency on the pumping and on the loss lead to gain saturation which govern the correlation volume in steady state.

These results clearly forbid the development of a photon bomb in lossy, finite, but amplifying random systems on the one hand, but also guarantee the coherence of the

emission. The relation between the atomic population inversion and the dynamics of the coherent part of the light field described by the Bethe-Salpeter-Equation constitutes the parameter-free link between microscopic and mesoscopic processes in random lasing which has been missing up to the present. The description of growth of the photon number density in both approaches establishes the selfconsistency chain that incorporates the full dynamics of the random laser. Hence the first parameter-free description of random lasing is established.

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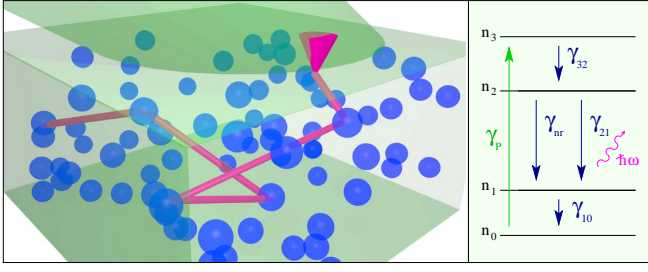


FIG. 1: **a** Random Laser setup. The pump diameter is assumed to be large compared to the measured emission. **b** Four level laser scheme.

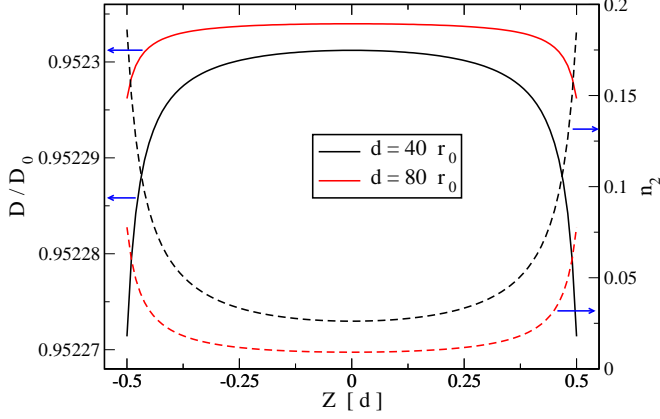


FIG. 2: The full and self-consistent diffusion constant D in units of the bare diffusion constant D_0 is shown as a function of the width Z across the sample for pump rate $P/\gamma_{21} = 2$. The sample width is set to be $d = 40r_0$ and $d = 80r_0$ respectively as indicated in the legend, where r_0 is the radius of the scatterers. As discussed in the text, the ratio D/D_0 remains practically unchanged, since the relative change observed in the graph is of the same order as the accuracy, involved in the numerical evaluation. This implies that interference effects do not change as a function of the sample width. The diffusion constant D can be seen to behave inversed compared to the inversion n_2 . Both quantities show a weak dependency to the sample's width.

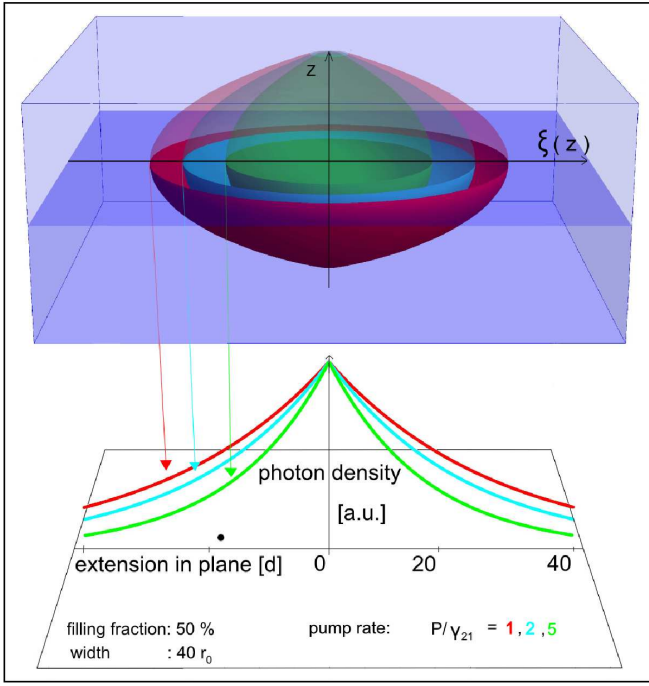


FIG. 3: **a** The correlation volume of the RL mode strongly decays with increasing pump strength. **b** Calculated photon density

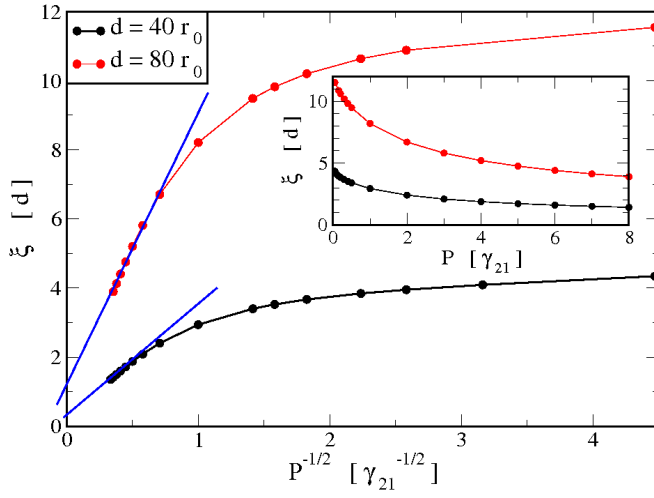


FIG. 4: Correlation length ξ as a function of the inverse square root of the pump rate, measured in units of the inverse square root of the transition rate γ_{21} , at the sample surface, i.e. $Z = 0.5d$.

The blue lines serve as a guide to the eye, emphasizing the linear behavior of the correlation length in this plot. This clearly reveals an inverse square root behavior of ξ as a function of pumping above threshold.